

Statistical mechanics of Floquet systems: the pervasive problem of near degeneracies

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Abstract

Although the statistical mechanics of periodically driven (“Floquet”) systems in contact with a heat bath has some formal analogy with the traditional statistical mechanics of undriven systems, closer examination reveals radical differences. In Floquet systems *all* quasienergies ε_j can be placed in a finite frequency interval $0 \leq \varepsilon_j < \omega$ (with ω the driving frequency and $\hbar = 1$), and the number of near degeneracies ($|\varepsilon_j - \varepsilon_k| \leq \delta$ for arbitrarily small δ) in this interval grows without limit as the dimension N of the Hilbert space increases. As we noted in a previous paper, this leads to pathologies, including drastic changes in the Floquet states, as N increases. In earlier work on Floquet systems in contact with a heat bath these difficulties were put aside by fixing N , while taking the coupling to the bath to be smaller than any quasienergy difference. This led to a simple explicit theory for the reduced density matrix, but with some major differences from the usual time independent statistical mechanics. We show that, for weak but finite coupling between system and heat bath, the accuracy of a calculation within the truncated Hilbert space spanned by the N lowest energy eigenstates of the undriven system is limited, as N increases indefinitely, only by the usual neglect of bath memory effects within the Born and Markov approximations. As we seek higher accuracy by increasing N , we inevitably encounter quasienergy differences *smaller* than the system-bath coupling. We therefore derive the steady state reduced density matrix without restriction on the size of quasienergy splittings. In general, it is no longer diagonal in the Floquet states. We analyze, in particular, the behavior near a weakly avoided crossing, where quasienergy near degeneracies routinely appear. The explicit form of our results for the density matrix gives a consistent prescription for the statistical mechanics for many periodically driven systems with N infinite, in spite of the Floquet state pathologies.

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I. INTRODUCTION

Time-periodic quantum systems are of great importance in several fields of science: atomic and solid-state systems driven by monochromatic electromagnetic fields [1]; quantum ratchets [2]; quantum chaos [3]. The Floquet theorem gives the solutions of the time-dependent Schrödinger equation with a time-periodic Hamiltonian (period $2\pi/\omega$) in the form $\exp(-i\varepsilon t)u(t)$, with $u(t)$ exhibiting the same periodicity as the Hamiltonian. The “quasienergy” ε plays a role analogous to the energy in static systems, determining in this case the behavior of the wave function under finite one-period, rather than infinitesimal, time translations. The quasienergy ε is defined only mod ω , and we follow the convention of taking $0 \leq \varepsilon < \omega$.

Real Floquet systems will ordinarily be in thermal contact with a bath. Energy is being continuously supplied by the periodic driving field, with relaxation provided by the bath. This leads to a fundamental question of statistical physics: Is there an asymptotic, time periodic, ensemble characterizing the quantum system and, if so, how does it depend on the coupling to and the temperature of the bath? For a time *independent* system with Hamiltonian H_0 the answer is given by the canonical reduced density operator $\rho = \exp(-\beta H_0)/Z$, where β is the inverse temperature, and Z is the partition function. This density operator is independent of the form and magnitude of the coupling to the environment, to lowest non-trivial order in that coupling, and the operator is diagonal in the energy eigenstates of the system Hamiltonian. Thus, even if two states are separated by an energy small compared to their coupling with the bath (“near degeneracy”), or even if they are degenerate, their statistical weights are given by their energy, and there is no anomaly associated with their near or strict degeneracy. That simplicity does *not* carry over to the case of near degeneracy in *quasienergy*.

For time-periodic systems in thermal contact with a bath the general long time behavior has not yet been fully clarified. In particular, the inevitable appearance of near degeneracies of quasienergies has not been addressed. Earlier work includes a number of papers [4, 5, 6, 7, 8, 9] which have discussed specific physical systems, treating either those whose quantum states span only a finite space, or others whose Hilbert space has been truncated to a finite set and then studied numerically, but always with coupling to the bath small with respect to any quasienergy spacing. Within such a finite state space, and in the limit of weak coupling

to the bath, a description of driven dissipative systems, as applied to driven Rydberg atoms in thermal contact with discrete wave guide modes, was introduced by Blümel *et al.* [4], and extended by Kohler *et al.* [7] to driven one-dimensional harmonic and anharmonic oscillators. An extension to strong coupling to the bath was presented by Thorwart *et al.* [10] for tunneling and relaxation of a particle in a bistable one-dimensional well, but using methods and approximations suitable only when a (small) finite number of system energy eigenstates is significantly accessible. A discussion of the statistical mechanics of a more general class of periodically driven systems was given by one of us [11], restricted to a finite Hilbert space with a non-degenerate quasienergy spectrum, in the limit of coupling to the bath much weaker than the smallest quasienergy spacing. In this limit, where the density operator is assumed diagonal in the representation of the Floquet states, the matrix elements can be obtained by solving a set of linear rate equations. In sharp contrast to the time independent case, they do depend on the explicit form (but not the magnitude) of the coupling between system and bath, and they are not simple smooth functions of the quasienergy.

In the general case of time-periodic quantum systems with a Hilbert space whose number of dimensions N increases without limit, the situation is more complex. Since the quasienergies are restricted to a finite range of width ω , the mean spacing between neighboring quasienergy levels approaches zero as their number increases. Therefore, any fixed coupling to the thermal environment is never small relative to all level spacings for a sufficiently large basis size. It becomes essential to deal with degenerate and near degenerate quasienergies, a situation which has been explicitly excluded in previous analyses. Indeed, in the limit $N \rightarrow \infty$ an infinite number of quasienergy degeneracies avoided by arbitrarily weak level repulsions may occur, and many anomalies of the spectrum and the Floquet states appear, as was pointed out in [12]. So it is of special importance to determine the effect of the bath when the coupling becomes comparable to or even large with respect to one or more of the relevant quasienergy splittings.

In this paper we address the critical question of the appropriate description of the steady state of time-periodic quantum systems with small but finite coupling to the bath. We review in Sec. II the derivation from the master equation of the set of linear equations that determine the reduced density matrix. This matrix is in general no longer diagonal in the basis of the Floquet states of the system, but it does approach a limiting time periodic form.

We obtain a set of linear homogeneous equations which determine all of the density matrix elements in the long time, stationary time-periodic state, Eq. (2.20). This result is no longer limited to the regime of bath coupling weak compared to all quasienergy spacings. For a finite Hilbert space and in the limit of infinitesimal coupling we discuss the differences from the conventional statistical mechanics of time-independent systems.

We next study in detail the influence of a single weakly avoided crossing of Floquet quasienergies (as a function of driving field strength) on a finite density matrix, since it is there that the near degeneracies of interest develop. In Sec. III we find that the important consequences of such an avoided crossing can be incorporated by introducing an additional effective coupling (3.4) between the two states involved, mediated by the bath. This coupling decreases with distance from the avoided crossing (which conveniently parameterizes the quasienergy separation of the two states), and it depends on the ratio of the coupling strength Γ of the two states to the bath, and their minimum quasienergy splitting, Δ . Remarkably, all of the diagonal elements of the density matrix in the representation of the “diabatic” states defined well away from the center of the avoided crossing, a basis which is fixed throughout the crossing, are determined by a set of linear rate equations (3.3) of exactly the structure familiar from ordinary time-independent statistical mechanics, but with this single added effective coupling rate. For $\Gamma \ll \Delta$ the additional coupling influences not only the (diagonal and off-diagonal) reduced density matrix elements for the two states themselves, but may completely change the whole steady state reduced density matrix, so that average system observables will in general change radically with driving field strength in the range of the avoided crossing. Physically, high order near resonances of the driving field between eigenstates of the undriven system can populate high energy levels, which then affect populations of lower energy levels in the relaxation cascade. In the opposite limit, $\Gamma \gg \Delta$, the coherence necessary for the high order resonance is disrupted by the interactions with the bath, and the avoided crossing has negligible influence on the reduced density matrix which, in this limit, is approximately diagonal in the diabatic basis. We present a simple, albeit somewhat artificial, example which can be solved in some detail, specifically exhibiting these effects.

We then use this insight to discuss time-periodic quantum systems with a Hilbert space of increasingly large dimension in Sec. IV. We find that weakly enough avoided crossings, while having great impact on the Floquet states, have no significant consequences for the

density matrix (in a λ -independent basis). It is this central result which justifies the neglect in many cases of all but a finite basis set in the calculation of the statistical mechanics of a Floquet system, as is essential for any numerical study. Specifically, for the calculation of statistical properties, we establish the validity of using the conventional approximation of eliminating from the *undriven* system spectrum all states of sufficiently high energy (the cutoff to be determined by the accuracy desired), and allowing the periodic driving field to mix only the remaining states. Within that finite basis set we have derived a prescription for calculating the full reduced density matrix, in general no longer diagonal in the Floquet representation. A brief summary is given in the conclusion, Sec. V.

II. REDUCED DENSITY MATRIX

A. Equations of motion and time periodic solutions

We review and extend the derivation of the steady state solution for the reduced density matrix in the weak-coupling limit (“Born-Markov approximation”) for Floquet systems, previously discussed [4, 7] only for situations where all quasienergy spacings were large compared with the coupling to the bath (and therefore only for systems restricted to a finite number of states). In contrast, we stress that the validity of the more general equations is not limited to that regime, and we give a detailed justification for an approximation using time-averaged rates.

We write the Hamiltonian generally as the sum of $H_s(t)$, describing the dynamics of the system driven by a time-periodic external field with period $2\pi/\omega$, the Hamiltonian H_b of the bath, and the interaction between system and bath H_{sb} ,

$$H(t) = H_s(t) + H_b + H_{sb}. \quad (2.1)$$

The total density operator of this closed coupled system, $\rho_T(t)$, is the solution of the equation of motion,

$$i\dot{\rho}_T(t) = [H(t), \rho_T(t)]. \quad (2.2)$$

We trace over bath variables to obtain the reduced density operator for the system,

$$\rho = \text{Tr}_b \rho_T, \quad (2.3)$$

and do the standard recasting of the master equation into an integro-differential equation (see, e.g., [14, 15]), whose iteration gives directly the expansion in powers of the system-bath interaction:

$$\frac{d}{dt}\tilde{\rho}(t) = - \int_0^t dt' \text{Tr}_b \left[\tilde{H}_{sb}(t), \left[\tilde{H}_{sb}(t'), \tilde{\rho}_T(t') \right] \right]. \quad (2.4)$$

Here the tilde labels operators in the interaction picture. We have omitted the linear term involving $\text{Tr}_b H_{sb}$, which ordinarily vanishes. This is the case, e.g., for an interaction between system and bath characterized by creation or destruction of single bath bosons, such as phonons or photons, or of free electrons or other fermions. Otherwise, this equation is still general and exact.

In order to make further progress, which depends on being able to trace over the bath variables under the integral, we make two essential simplifications: (i) second order perturbation theory [16] in H_{sb} and (ii) the Markov approximation. Their validity relies ultimately on being able to neglect any memory effects within the bath. The correlation times associated with the bath must be small compared with the times characterizing the evolution of the reduced density operator (see the condition (2.10) below). First, to lowest non-vanishing order in H_{sb} we take the total density operator $\rho_T(t')$ under the integral as a simple product of system and bath factors,

$$\tilde{\rho}_T(t') \approx \tilde{\rho}(t') \rho_b. \quad (2.5)$$

The physical meaning of this approximation is that we neglect more than a single interaction of the system with the bath during the bath correlation time. The effect of higher order correlations, where subsequent interactions depend on the state of the bath modified by earlier ones, is negligible, as we show in more detail in Appendix A.

We make the specific, but inessential, choice of coupling Hamiltonian:

$$H_{sb} = \gamma AB, \quad (2.6)$$

where the Hermitian operator A acts only on the Floquet system, and the Hermitian operator B acts only on the heat bath. A sum of such factorized operator products, $\gamma \sum_\alpha A_\alpha B_\alpha$, is also readily treated. But if the various bath operators B_α are dynamically uncorrelated with each other, then the results below are modified only by an extra index on each matrix element of A and each spectral function $g(E)$ (to be defined below), and a sum over that index. We take the single product to avoid the consequent algebraic clutter, since no essential

difference results in the ultimate equations (the extra sums being merely incorporated into the definition of the complex “rates” below (2.14)). Then the trace over bath variables involves only the single combination

$$G(\tau) = \text{Tr}_b \rho_b \tilde{B}(t) \tilde{B}(t - \tau), \quad (2.7)$$

This bath correlation function is characterized by a decay time τ_c , which we assume to be much shorter than the times characterizing the evolution of the reduced density operator $\tilde{\rho}(t)$ (see Eq. (2.10) below). Then the main contribution to the integral in (2.4) is from values of t' within a range of order τ_c below the upper limit t . Thus to a good approximation we can extend the lower limit to $-\infty$ and also make the Markov approximation: replace t' by t in the argument of $\tilde{\rho}_T$ under the integral. Returning to the Schrödinger picture and changing the integration variable to $\tau = t - t'$, we then have

$$\frac{d\rho}{dt} + i[H_s(t), \rho(t)] = -\gamma^2 \int_0^\infty d\tau \left\{ G(\tau) \left[A \tilde{A}(t - \tau, t) \rho(t) - \tilde{A}(t - \tau, t) \rho(t) A \right] + \text{H.c.} \right\}. \quad (2.8)$$

The rate of change of the reduced density operator due to the coupling to the bath is of order

$$\Gamma_2 = \gamma^2 \tau_c \langle A^2 \rangle \langle B^2 \rangle, \quad (2.9)$$

suggesting that the condition for validity of the “Born” and Markov approximations is

$$\tau_c \Gamma_2 \ll 1. \quad (2.10)$$

Appendix A demonstrates that this same inequality assures that higher order corrections are negligible. The validity of the above approximations is discussed in the literature on undriven systems; a particularly clear discussion is given in Ref. [15]. As the approximations have been made within the interaction picture, they are equally valid for a time-periodic system Hamiltonian $H_s(t)$. We emphasize that we do *not* require infinitesimal coupling to the bath. That coupling need only be small enough to satisfy the condition (2.10).

We recall that the solutions of the isolated system Schrödinger equation $i\partial_t|\phi\rangle = H_s(t)|\phi\rangle$ are of the Floquet form $|\phi_j(t)\rangle = \exp(-i\varepsilon_j t)|u_j(t)\rangle$ with the quasienergies ε_j chosen to lie in the strip $0 \leq \varepsilon < \omega$ and with the time-periodic parts $|u_j(t)\rangle = |u_j(t + 2\pi/\omega)\rangle$ forming a complete orthonormal basis for the system *at any given time* t . We introduce the matrix elements of the reduced density operator in this time-periodic basis,

$$\rho_{ij}(t) = \langle u_i(t) | \rho(t) | u_j(t) \rangle, \quad (2.11)$$

which will turn out to be very convenient in the following calculations and which, as we will show later, leads to time-independent density matrix elements in steady state. In this basis the equation of motion of the reduced density matrix elements takes the form

$$\begin{aligned} \left(\frac{\partial}{\partial t} + i\varepsilon_{ij}\right)\rho_{ij}(t) = & -\gamma^2 \sum_{k,l,M,m} e^{i(M+m)\omega t} \left[+ A_{ik}(M)A_{kl}(m)\rho_{lj}(t) \int_0^\infty d\tau G(\tau)e^{i(\varepsilon_{lk}-m\omega)\tau} \right. \\ & - A_{ik}(m)\rho_{kl}(t)A_{lj}(M) \int_0^\infty d\tau G(\tau)e^{i(\varepsilon_{ki}-m\omega)\tau} \\ & + \rho_{ik}(t)A_{kl}(m)A_{lj}(M) \int_0^\infty d\tau G^*(\tau)e^{i(\varepsilon_{lk}-m\omega)\tau} \\ & \left. - A_{ik}(M)\rho_{kl}(t)A_{lj}(m) \int_0^\infty d\tau G^*(\tau)e^{i(\varepsilon_{jl}-m\omega)\tau} \right], \end{aligned} \quad (2.12)$$

where we defined the Fourier components of the matrix elements of the system operator A ,

$$\langle u_i(t)|A|u_j(t)\rangle \equiv \sum_m A_{ij}(m)e^{im\omega t}, \quad (2.13)$$

with the property $A_{kl}(m) = A_{lk}^*(-m)$, and where we have introduced the notation $\varepsilon_{ij} \equiv \varepsilon_i - \varepsilon_j$.

We simplify the notation by introducing complex time-periodic “rates”

$$R_{ij;kl}(t) = \sum_K e^{iK\omega t} R_{ij;kl}(K), \quad (2.14)$$

with Fourier coefficients

$$R_{ij;kl}(K) = \sum_m R_{ij;kl}^m(K) = 2\pi\gamma^2 \sum_m A_{ij}(m+K)A_{kl}^*(m)g(\varepsilon_{lk} - m\omega), \quad (2.15)$$

where $g(E)$ is the Fourier transform of $G(\tau)$. We will approximate

$$\int_0^\infty d\tau G(\tau)e^{-iE\tau} \approx \pi g(E), \quad (2.16)$$

neglecting a principal value contribution. From the definition of $G(\tau)$, Eq. (2.7), one finds

$$g(E) = \sum_{\nu,\nu'} P_\nu |\langle \nu|B|\nu'\rangle|^2 \delta(E + E_{\nu'} - E_\nu), \quad (2.17)$$

where ν labels bath states of energy E_ν which are in thermal equilibrium and have occupation $P_\nu \propto \exp(-\beta E_\nu)$.

Then we have as the fundamental equation of motion of the reduced density matrix elements in the Floquet representation, the following set of linear differential equations:

$$\left(\frac{\partial}{\partial t} + i\varepsilon_{ij}\right)\rho_{ij}(t) = -\frac{1}{2} \sum_{k,l} \left[\rho_{lj}(t)R_{ik;lk}(t) + \rho_{il}(t)R_{jk;lk}^*(t) - \rho_{kl}(t)(R_{lj;ki}(t) + R_{ki;lj}^*(t)) \right]. \quad (2.18)$$

This is the generalized Master equation for all density matrix elements. We next replace the time periodic rates $R_{lj;ki}(t)$ by their average over one driving period,

$$R_{lj;ki} = \sum_m R_{lj;ki}^m = R_{lj;ki}(K = 0). \quad (2.19)$$

This approximation [7] implicitly assumes that the density matrix elements do not vary substantially over a period of the driving, and it is further discussed in Appendix B. We then find a steady state solution of (2.18) for which the matrix elements ρ_{ij} are independent of time:

$$i\varepsilon_{ij}\rho_{ij} = -\frac{1}{2}\sum_{k,l}\left[\rho_{lj}R_{ik;lk} + \rho_{il}R_{jk;lk}^* - \rho_{kl}(R_{lj;ki} + R_{ki;lj}^*)\right]. \quad (2.20)$$

B. Large quasienergy splittings

The states of some systems of interest span only a finite dimensional space (e.g., spin systems). Because of the finite number of states, at a given driving strength λ there will be a smallest quasienergy separation. In this subsection we take that smallest separation to be large compared to the coupling to the heat bath (an assumption made, in fact, in all earlier work on driven dissipative systems in the weak-coupling limit), and we will point to both the similarities to and the differences from the conventional statistical mechanics of time-independent systems.

In the limit where the absolute values of all “rates” $|R_{ij;kl}|$ are sufficiently small compared with *all* splittings $|\varepsilon_{ij}|$, it is clear from (2.20) that all off-diagonal density matrix elements can be neglected, and we arrive at the familiar form,

$$0 = \rho_{ii}\sum_k R_{ik} - \sum_k \rho_{kk}R_{ki}, \quad (2.21)$$

relating the state occupation probabilities $p_i \equiv \rho_{ii}$. Here the diagonal time-independent rates,

$$R_{ki} = R_{ki;ki} = 2\pi\gamma^2\sum_m |A_{ki}(m)|^2 g(\varepsilon_{ik} - m\omega), \quad (2.22)$$

are of the form of the usual transition rates of perturbation theory, the square of coupling matrix elements times the density of states (at the energy transferred from bath to system), summed over initial states of the bath with appropriate thermal equilibrium weighting. Of course, in the static case the equation also holds without the summation over k (detailed balance). That is *not* the case here.

As was pointed out earlier [9, 11], there are some important differences in this result from that familiar in static problems. Because of the possibility of absorption and emission of multiple quanta of the time-dependent driving field, the occupation probabilities are not simple Boltzmann factors, and indeed they depend on the detailed form of the Hamiltonian coupling the system to the heat bath. From the definition (2.17) of the spectrum of the bath correlation function we have

$$g(E) = g(-E)e^{-\beta E}, \quad (2.23)$$

which is used in the time-independent case to derive the detailed balance equation. In a Floquet system it leads, using (2.17) and (2.22), to the relation

$$R_{jj'}^m = R_{j'j}^{-m} e^{\beta(\varepsilon_j - \varepsilon_{j'} + m\omega)}, \quad (2.24)$$

relating only the pieces of the transition rates between system states j and j' which involve the transfer of exactly m quanta from the field. After summation over m we find for the ratio of the full transition rates between these states,

$$\frac{R_{jj'}}{R_{j'j}} = \frac{\sum_m R_{jj'}^m}{e^{-\beta(\varepsilon_j - \varepsilon_{j'})} \sum_m R_{jj'}^m e^{-\beta m\omega}}. \quad (2.25)$$

There are situations where the $R_{jj'}^m$ have for some pairs (j, j') just one value of m , say m_0 , with a non-negligible value. This is the case for a very small driving amplitude ($\lambda \ll 1$) or for high-lying states of the undriven system that are weakly affected by the periodic driving, if the levels j and j' are far enough from any avoided crossing. Then in the above ratio the terms $R_{jj'}^{m_0}$ cancel and the form of the coupling Hamiltonian H_{sb} is no longer relevant. One gets $R_{jj'}/R_{j'j} \approx \exp[\beta(\varepsilon_j - \varepsilon_{j'} + m_0\omega)]$, much as in the time independent case, but reflecting the need for m_0 quanta of the driving field.

In general, the states are mixed by the driving field, giving rates $\sum_m R_{jj'}^m$, where more than one value of m contributes significantly. Consequently the above ratio $R_{jj'}/R_{j'j}$ will no longer depend just on a single quasienergy difference. In order to find the stationary weights $p_j \equiv \rho_{jj}$ one always has to solve Eq. (2.21). This solution will in general not fulfill the detailed balance equations. Even for the simple situation above, where the direct and inverse rates for states j and j' have a simple Boltzmann factor ratio, we can not then conclude that the corresponding probabilities p_j and $p_{j'}$ have the same ratio. As in the case of a time independent Hamiltonian, the stationary solution of Eq. (2.21) will not depend on the overall amplitude γ of the coupling Hamiltonian (if γ is sufficiently small for lowest

order perturbation theory to be valid). In contrast to the time-independent case, however, the solution *will* depend on the rates $R_{jj'}^m$, and thus on the precise form of the coupling Hamiltonian H_{sb} , as noted in [11].

III. ISOLATED WEAKLY AVOIDED CROSSING

As a function of the strength of the driving field λ the quasienergy spectrum of Floquet systems is generally pervaded by weakly avoided crossings, near which quasienergy separations are small and the Floquet states are rapidly varying. These features can have important consequences for the density matrix. We consider a finite set of Floquet states for which, in the neighborhood of the driving field strength λ being studied, there is a single weakly avoided crossing, with small quasienergy splitting Δ at the center of the crossing at λ_0 near λ . At the same time all other quasienergy separations at λ are assumed to be large compared to the appropriate measure of the coupling strength to the bath. Then only the two Floquet states involved in the weakly avoided crossing are sensitive functions of λ over the small crossing region. For very weak coupling to the bath compared with Δ we know from the results of Sec. IIB that ρ is diagonal in the basis of those Floquet states. In contrast to the time independent case, where the density matrix elements depend only on the state energies, here it will change rapidly as the Floquet states do, not only within the subspace of the states of the avoided crossing but, as we will show, in general throughout the rest of the density matrix, as well. In the opposite limit, when the system-bath interaction effectively broadens those two states by much more than Δ , we will show that there is little influence from the crossing on the density matrix (in a λ -independent basis) or on the statistical properties of the system determined therefrom. This will allow us in the next section to extend our analysis to many driven systems for which the quasienergy spectrum is dense, as is often the case in practice.

We review briefly the description of the states and quasienergies in the neighborhood of such a weakly avoided crossing, introducing the notation which will be used later. We assume that within a suitably small neighborhood of driving strengths around λ_0 the time-periodic parts $|u_1(t)\rangle$ and $|u_2(t)\rangle$ of the unperturbed “adiabatic” states (i.e., if there were no avoided crossing) do not depend significantly on λ , and that the unperturbed quasienergies are locally linear in λ with slopes σ_1 and σ_2 , respectively. Then, the quasienergies and the

time-periodic part of the Floquet states can be well described by the results of diagonalizing a 2×2 Hamiltonian matrix (the Floquet analog of nearly degenerate perturbation theory, using the extension of Rayleigh-Schrödinger perturbation theory developed by Sambé [17]). The quasienergies are given by

$$\varepsilon_{a,b} = \varepsilon_0 + \frac{(\sigma_1 + \sigma_2)}{2}(\lambda - \lambda_0) \pm \frac{1}{2}\sqrt{(\sigma_1 - \sigma_2)^2(\lambda - \lambda_0)^2 + \Delta^2}, \quad (3.1)$$

where the indices a, b refer to the two “adiabatic” Floquet states continuous in λ , and Δ is the minimum quasienergy splitting, at the center of the crossing, where $\lambda = \lambda_0$. This is shown schematically in Fig. 1. The adiabatic states a and b exchange their character when λ is varied, while by assumption the diabatic states 1 and 2 remain unchanged.

We define the dimensionless distance d from the avoided crossing at $\lambda = \lambda_0$ as

$$d \equiv (\lambda - \lambda_0)(\sigma_1 - \sigma_2)/\Delta, \quad (3.2)$$

the ratio of the quasienergy separation of the unperturbed states at λ to the gap Δ . The central region of the avoided crossing is thus given by $|d| < 1$.

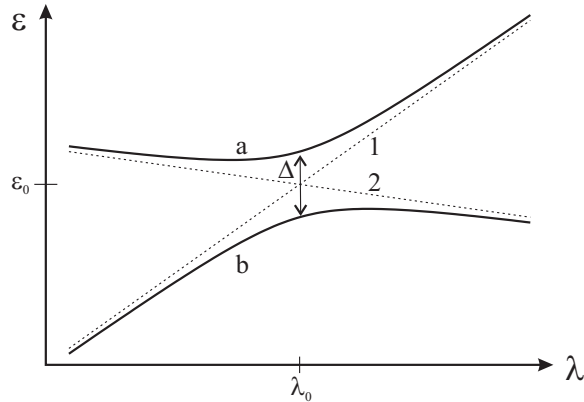


FIG. 1: Behavior of quasienergies around an avoided crossing of size Δ . The dashed lines show diabatic (unmixed) states 1, 2 and the solid curves the adiabatic Floquet states a, b .

A. Diabatic basis and effective rate R^{ac}

Over the narrow range of the weakly avoided crossing the diabatic states $|u_1(t)\rangle$ and $|u_2(t)\rangle$, as well as the time periodic parts of all other Floquet states, are approximately

independent of λ , so they constitute a natural fixed basis of time periodic functions to use in studying this region. In Appendix C we derive the set of rate equations for the reduced density matrix elements ρ_{ij} in this diabatic basis. Quite strikingly, under a number of plausible assumptions, they allow for a separate determination of the diagonal and off-diagonal elements. The former are simply given by

$$0 = \rho_{ii} \sum_j \bar{R}_{ij} - \sum_j \rho_{jj} \bar{R}_{ji}, \quad (3.3)$$

where the rates designated with a bar, \bar{R}_{ij} , are defined in the diabatic basis in analogy to Eq. (2.22), except for

$$\bar{R}_{12} = \bar{R}_{21} = R^{\text{ac}} \equiv \frac{\Gamma}{4d^2 + (\Gamma/\Delta)^2}, \quad (3.4)$$

with the superscript on R^{ac} a reminder that this is an effective rate associated with the small quasienergy separation near the avoided crossing (ac). Here we have introduced the symbol

$$\Gamma \equiv \sum'_k (\bar{R}_{1k} + \bar{R}_{2k}) + \bar{R}_{11} + \bar{R}_{22} - 2\bar{R}_{11;22}, \quad (3.5)$$

where the prime on the summation denotes the restriction $k \neq 1, 2$. (Note that the sum of the last 3 terms is positive, in spite of the minus sign; by definition they can be written $2\pi\gamma^2 \sum_m [A_{11}(m) - A_{22}(m)]^2 g(-m\omega)$.) The rate Γ will turn out to be the relevant coupling to the heat bath, specific to the crossing of states 1 and 2, that will be the measure of whether that coupling can be considered weak or strong relative to the quasienergy splitting of those states.

These equations (3.3) for the diagonal density matrix elements ρ_{ij} in the diabatic basis have exactly the formal structure of the corresponding equations in the limit of bath coupling small compared to all quasienergy splittings, Eq. (2.21). The results differ from that simpler situation in three essential ways:

(i) The effective rates R^{ac} between the two special states 1 and 2 are not simply the second order rates imposed directly by the bath. They reflect the mixing in a more complex way, effectively including the driving induced coupling to all orders, and they can be simply determined from the rate Γ defined in Eq. (3.5) and the properties of the avoided crossing, namely the distance d and the minimal quasienergy splitting Δ .

(ii) The dependence on the parameter λ , which in the original adiabatic basis strongly affects a great number of rates, appears in the diabatic basis only in the single rate R^{ac} (see Eq. (3.4), where the distance from the avoided crossing, d , is linearly related to λ).

(iii) These are not the *only* density matrix elements; there exist also two non-vanishing off-diagonal elements ρ_{12} and $\rho_{21} = \rho_{12}^*$ in this diabatic basis, given by

$$\rho_{12} = \frac{\rho_{11} - \rho_{22}}{2d - i\Gamma/\Delta}. \quad (3.6)$$

Their importance can be assessed by a comparison of the absolute value of ρ_{12} to the sum of the positive and real values ρ_{11} and ρ_{22} :

$$\frac{|\rho_{12}|}{\rho_{11} + \rho_{22}} = \frac{|\rho_{11} - \rho_{22}|}{\rho_{11} + \rho_{22}} \frac{1}{\sqrt{4d^2 + (\Gamma/\Delta)^2}} \leq \frac{\Delta}{\Gamma}, \quad (3.7)$$

which is negligible in the limit $\Gamma \gg \Delta$, (that is, where the coupling to the heat bath is stronger than the minimum quasienergy separation of the Floquet states). This demonstrates that in this limit the density matrix in this two-dimensional subspace is approximately diagonal in the diabatic basis. In the opposite limit, the density matrix in the subspace is nearly diagonal in the adiabatic Floquet basis. The appropriate basis to diagonalize ρ is given in general in Appendix D.

Although we have no general analytic solution of the equations (3.3) for the diagonal density matrix elements, we can say a good deal about the consequences of a weakly avoided crossing, as the relevance of the effective rate R^{ac} depends crucially on the ratio of the bath-induced rate Γ to the size of the avoided crossing Δ .

1. $\Gamma \ll \Delta$

In this limit (which is just the case discussed in Sec. IIB above) the rate Γ characterizing the coupling to the heat bath is even smaller than the quasienergy splitting at the weakly avoided crossing. Consequently, the effective rate R^{ac} , Eq. (3.5), is much larger than Γ in the center of that crossing,

$$R^{\text{ac}}(d = 0) \gg \Gamma. \quad (3.8)$$

The immediate consequence of this large effective rate is that the diagonal density matrix elements $\rho_{11} \approx \rho_{22}$ become equal there, which may drastically change all other weights ρ_{jj} , coupled directly or indirectly via Eq. (3.3) to ρ_{11} or ρ_{22} , relative to the corresponding values just outside the avoided crossing. An explicit example demonstrating such a drastic change for the entire density matrix is studied in Sec. IIIB.

The off-diagonal matrix element ρ_{12} is not necessarily small, though, since it is given by the product of one large and one small factor; see Eq. (3.6). In fact, as discussed in Appendix D, the off-diagonal element is negligible in the adiabatic basis only.

2. $\Gamma \gg \Delta$

In this limit the rate Γ induced by the coupling to the heat bath is stronger than the size of the weakly avoided crossing. Consequently, the effective rate R^{ac} , Eq. (3.5), is much smaller than Γ throughout the whole range of the avoided crossing,

$$R^{\text{ac}} \ll \Gamma. \quad (3.9)$$

This suggests that the effective rate R^{ac} due to the avoided crossing has negligible influence compared to the other rates affecting states 1 and 2. Therefore the entire density matrix is not affected by the existence of the weakly avoided crossing. The off-diagonal matrix element ρ_{12} is negligible in this limit.

There is an exception to the above conclusion if the two sums $\sum'_k \bar{R}_{1k}$ and $\sum'_k \bar{R}_{2k}$ contributing to Γ have vastly different magnitudes. This is possible, as we are explicitly looking at a very weakly avoided crossing, and this typically arises from such dissimilar pairs of states, such as those arising from undriven states one of which is of low and the other of very high energy. In what follows we will take state 2 to be the one with higher average energy $\langle H_s \rangle$. There are two cases:

- $\sum'_k \bar{R}_{2k} \ll R^{\text{ac}} \ll \sum'_k \bar{R}_{1k}$: This case may occur if the heat bath is dominated by phonons (or by other bosons) with an upper energy limit ω_D , small compared to the separation in average energy of the diabatic state 2 and its neighbors, so that state 2 will decay primarily to state 1, with flux $\rho_{22}R^{\text{ac}}$. Similarly, state 2 is fed primarily by the direct process from state 1 with flux $\rho_{11}R^{\text{ac}}$ (in the extreme limit of all $\bar{R}_{j2} = 0$, *only* by this process). Since stationarity demands that the incoming and the outgoing flow for state 2 are equal, one finds $\rho_{11} \approx \rho_{22}$, with corresponding large changes in other state occupations ρ_{jj} . This modification of the density matrix occurs for all values of d and Γ/Δ for which the presumed rate inequalities hold. Values of those parameters can be significantly larger than unity, but eventually large enough values will reduce R^{ac} to the point where the lower inequality is violated.

- $\sum'_k \bar{R}_{1k} \ll R^{\text{ac}} \ll \sum'_k \bar{R}_{2k}$: This case may occur when the heat bath is dominated by the electromagnetic radiation field, with photons of arbitrarily high energy and large density of states. Then state 2, essentially a high energy undriven state, will decay rapidly. By precisely the same argument used in the previous case, $\rho_{11} \approx \rho_{22}$, and states now strongly connected to 2 acquire substantial occupancy. Again, the ranges of d and Γ/Δ over which this situation holds can be large.

B. A simple example

It is instructive to obtain specific results for a simple, albeit somewhat artificial, example of a system. We assume first that for Floquet states labeled by some index, $n = 1, 2, \dots, N$, the thermal bath leads to coupling only of neighboring states — i.e., the only non-zero rates are of the form $R_{n,n\pm 1}$, as shown in Fig. 2, and that we are in the limit of coupling to the bath weak with respect to all quasienergy differences. In this special situation the lowest state 1 has to be in detailed balance with state 2 and from this it follows that all states are in detailed balance. If we make the specific choice $R_{n,n-1} = R_d$ and $R_{n,n+1} = R_u$, each independent of n , and denote the ratio $R_u/R_d \equiv \gamma < 1$, then the occupations $p_n \equiv \rho_{nn} = (1 - \gamma)\gamma^{n-1}/(1 - \gamma^N)$ fall off exponentially with the index n . We assume N to be sufficiently large that we can neglect (for algebraic simplicity) the factor γ^N below.

Now we want to study how these occupations change if there is a single weakly avoided crossing of states n_1 and n_2 (in the previous section these were the states called 1 and 2). This introduces the additional rate R^{ac} . It simplifies the algebra to define the fluxes between states n and $n - 1$,

$$F_{n,n-1} \equiv p_n R_{n,n-1} - p_{n-1} R_{n-1,n}, \quad (3.10)$$

in terms of which we can rewrite the basic equations derived above for the diagonal density matrix elements, (3.3), as

$$(p_{n_1} - p_{n_2})R^{\text{ac}} = F_{n_2,n_2-1} - F_{n_2+1,n_2} \quad (3.11)$$

$$(p_{n_1} - p_{n_2})R^{\text{ac}} = F_{n_1+1,n_1} - F_{n_1,n_1-1} \quad (3.12)$$

$$0 = F_{n+1,n} - F_{n,n-1} \quad \text{for} \quad n \neq n_1, n_2 \quad \text{and} \quad n \neq 1 \quad (3.13)$$

$$0 = F_{2,1}. \quad (3.14)$$

Their solution is

$$F_{n,n-1} = \begin{cases} 0 & ; n \leq n_1 \\ F & ; n_1 < n \leq n_2 \\ 0 & ; n > n_2 \end{cases} , \quad (3.15)$$

with

$$F = (p_{n_1} - p_{n_2})R^{\text{ac}}, \quad (3.16)$$

being the flux from n_1 to n_2 due to the avoided crossing. This result is illustrated in Fig. 2. The states above n_2 and the states below n_1 are still in detailed balance — i.e. $F_{n,n-1} = 0$, as without the avoided crossing. The states from n_1 to n_2 , however, are no longer in detailed balance and there is a stationary flux F connecting them, due to the avoided crossing.

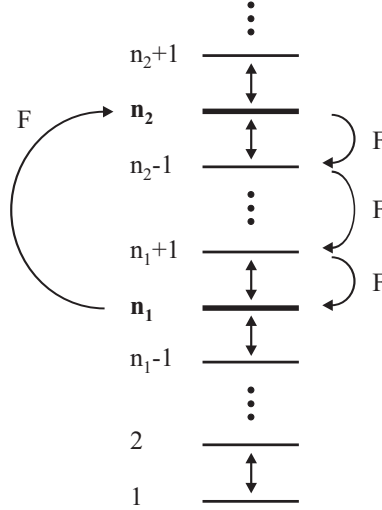


FIG. 2: Simple example with a thermal bath coupling neighboring states only (double headed arrows) and flux F introduced by the effective rate R^{ac} due to an avoided crossing of states n_1 and n_2 .

For the occupations p_n we find

$$p_n = \begin{cases} p_1 \gamma^{n-1} & ; n \leq n_1 \\ p_{n_1} \frac{r + \gamma^{n-n_1}}{r+1} & ; n_1 < n \leq n_2 \\ p_{n_2} \gamma^{n-n_2} & ; n > n_2 \end{cases} , \quad (3.17)$$

with

$$r \equiv \frac{R^{\text{ac}}}{R_d} \frac{1 - \gamma^{n_2 - n_1}}{1 - \gamma}, \quad (3.18)$$

and p_1 determined by the normalization $\sum_n p_n = 1$.

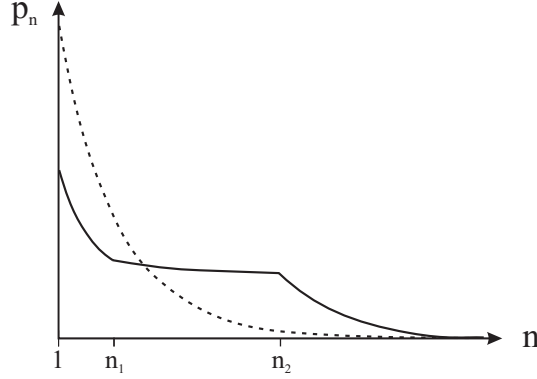


FIG. 3: Dramatic change in occupations p_n with (solid curve) and without (dashed curve) avoided crossing.

The parameter r measures the ratio of the effective rate R^{ac} describing the direct connection of the state n_2 to the state with which it is crossing, namely n_1 , relative to its rate of loss R_d to the state directly below it, times a factor of order one. There are three distinct interesting ranges of r in terms of the behavior of the density matrix elements:

(i) When $r \gg 1$ we have $p_{n_2} \approx p_{n_1}$ and almost constant weights between n_1 and n_2 . The weights of states above n_2 are much larger than they would be without the avoided crossing, due to the drastically increased weight p_{n_2} , which is being fed directly by n_1 .

(ii) For $\gamma^{n_2 - n_1} \ll r \ll 1$ we have $p_{n_2} \approx p_{n_1} r$, still a much higher occupancy than in the absence of the avoided crossing. Again the weights of states above n_2 are much larger than they would be without the avoided crossing. This case, with r only slightly less than 1, is sketched in Fig. 3.

(iii) If r decreases still further, $r \ll \gamma^{n_2 - n_1}$, then $p_n \approx p_1 \gamma^{n-1}$, which is the result without the avoided crossing. Equilibration with the bath will be completely dominated by the transition processes along the ladder of states.

This model admittedly has some special features which make it soluble. For instance, there is detailed balance in the absence of an avoided crossing. But it illustrates the dramatic change in all occupations due to a single avoided crossing.

IV. LIMIT OF INFINITE DIMENSIONAL HILBERT SPACE

As we have already noted, there are some interesting Floquet systems, such as finite spin systems, which are limited to a space spanned by a finite number of basis states. Their statistical mechanics can then be calculated (numerically) from the equations and analysis of the previous sections of this paper (and we will not be concerned with them further here). However, many others, like a particle confined by a potential well, have eigenstates spanning infinite Hilbert spaces, with a quasienergy spectrum which becomes dense and Floquet states which change rapidly as the basis size N increases, so that the above results do not readily apply. In fact, our results also permit the calculation to any desired accuracy of many of these, as well, by appropriate truncation of the basis set, as we now show.

We limit ourselves to systems with an unbounded discrete spectrum of *undriven* energy eigenstates. We take the truncated Hilbert space spanned by the N lowest energy eigenstates, the usual choice made in numerical calculations. Within this approximation the Floquet states of the isolated system (at any fixed time) are confined to that finite dimensional space. For the undriven system the statistical (Boltzmann) weights of the states neglected by the truncation can be made arbitrarily small by a suitably large choice of the energy cutoff. We now increase the Hilbert space accessible to the driven system by the addition of the $(N+1)$ st undriven energy eigenstate. With increasing λ , as its quasienergy nears that of one of the original N Floquet states, it will mix significantly with that state. But we have shown above (section III A 2) that, if the corresponding avoided crossing is sufficiently weak, $\Delta \ll \Gamma$, then the reduced density matrix is approximately diagonal in the unmixed diabatic states, and the addition of the new state to the basis set remains of little statistical significance. Therefore, if we can demonstrate that all of the states neglected by the truncation of high energy undriven eigenstates mix with those retained only through such weakly avoided crossings, then we can choose the energy cutoff of the basis to obtain statistical properties to any desired accuracy.

Indeed, this desired behavior of the quasienergy of a Floquet state arising from a high energy undriven eigenstate has been seen in numerical calculations [1]. Such a state is found to mix with the Floquet states that arise from low energy undriven eigenstates, as λ increases, only near weakly avoided crossings. In the appendix of Ref. [12] we have analytically demonstrated the effect explicitly, with an exponential decrease with basis size

N of newly introduced quasienergy gaps, for systems whose undriven energy eigenvalues above some level exhibit increasing spacings between successive levels (such as a particle confined by a one-dimensional potential which increases faster than harmonically). This can be rationalized in terms of a perturbation expansion: an increasingly large number of driving field quanta is needed to connect to higher lying states, which are modified only in increasingly higher order perturbation theory. More generally, numerical studies [13] suggest that once the first N undriven states have been included in the basis, the Floquet states arising from the first N' of these, for suitably large $N - N'$, are affected by further extension of the basis set only by weakly avoided crossings. Again, perturbation theory suggests that this is a reflection of the very different spatial behavior of these states.

For a class of periodically driven systems in an infinite Hilbert space we earlier found [12] many anomalies due to the infinite number of ever more weakly avoided crossings. The present results show that these anomalies, and the non-convergence of the Floquet states with increasing basis size, lead to negligible consequences for the reduced density operator, or for the statistical properties calculated therefrom, when the system is in *fixed* weak contact with a thermal bath.

In practice, one has to determine the quasienergy spectrum for increasing basis size N until (i) newly appearing avoided crossings are small enough, $\Delta \ll \Gamma$, and (ii) the statistical (Boltzmann) weight of the newly introduced states, plus that of all neglected states, is small enough to achieve the desired accuracy. Once the basis size N is appropriately chosen, one can use Eq. (2.20) to determine the density matrix elements and calculate statistical properties.

It is important to recognize that, because of the requirement $\Delta \ll \Gamma$ for avoided crossings with all neglected basis states, the minimum truncation size N depends in general on the *magnitude*, as well as the form, of the system-bath coupling, since both enter the determination of Γ . Indeed, the full statistical properties of the Floquet system will depend on that magnitude (we have already emphasized in Sec. IIB the general dependence on the *form* of the coupling), even if the coupling is very weak: For a given magnitude we choose N to achieve a desired accuracy. We now reduce the magnitude (keeping the form fixed) sufficiently that Γ becomes less than one of the gaps Δ associated with an avoided crossing of a state neglected by the truncation with a state that has been retained. Then that avoided crossing becomes no longer irrelevant. It leads to a large rate R^{ac} , Eq. (3.4), that

may cause quite different statistical weights, as was explicitly shown for the simple example in Sec. III B. We can understand this as the appearance of a new, high order, near resonance which is no longer disrupted by the bath interaction, resulting in the appreciable occupation of the upper (higher average energy) level of that pair, and modifying the statistical weight of the states to which it is connected.

This is emphatically not to suggest the lack of a well-defined statistical behavior of a Floquet system with given coupling to a bath. Indeed, the above procedure describes how that is to be calculated to any desired accuracy. However, the behavior does depend on the specific magnitude and form of that coupling, in sharp contrast to the situation for conventional time-independent statistical mechanics.

V. CONCLUSION

We have derived the general equations (2.20) for the reduced density matrix, in the stationary state, of Floquet systems with weak but *finite* coupling to a heat bath, previously discussed [4, 7, 9, 11] only for situations where all quasienergy spacings were large compared with the coupling to the bath (and therefore only for systems restricted to a finite number of states). New and interesting results give a prescription for practical calculation of the steady state statistical properties of many realistic Floquet systems, including ones with dense quasienergy spectra.

If the coupling to the bath Γ is restricted only to be small compared to the driving frequency ω and the inverse bath relaxation time $1/\tau_c$, the diagonal and off-diagonal matrix elements are approximately time-independent in the basis of the time-periodic parts of the Floquet states. In the special case of a Hilbert space truncated to finite dimensions and the coupling weak compared to all quasienergy separations, then the equations simplify and are equivalent to those discussed in [11] and [9]. The density matrix is diagonal, but the values of the diagonal elements (occupation probabilities of the Floquet states) depend on the detailed form (though not on the magnitude, as long as it is sufficiently weak) of the coupling of the system to the bath.

Weakly avoided crossings, where quasienergies become nearly degenerate, are a pervasive feature of Floquet systems. Therefore, we have analyzed in detail the general density matrix equations in such regions. The rate equations for diagonal density matrix elements in the

diabatic basis, Eq. (3.3), are shown to be identical to those well away from the crossing and with arbitrarily weak coupling to the heat bath, except that the avoided crossing introduces a new effective rate R^{ac} , defined in Eq. (3.4), between the diabatic states 1 and 2 of the crossing. This rate depends on the effective coupling Γ of the states 1 and 2 to the heat bath, the distance d from the avoided crossing, and the relative strength of the coupling to the heat bath, Γ/Δ . In addition, significant off-diagonal density matrix elements ρ_{12} and ρ_{21} may appear (see Eq. (3.6)).

The density operator changes drastically with λ in the neighborhood of an avoided crossing, $d \lesssim 1$, if also the bath coupling is weak enough, $\Gamma \lesssim \Delta$. Quite importantly, significant changes occur in the weights not only of the states mixed in the crossing, but in those of many other states, as well. At the center of the crossing the two states of the crossing pair acquire equal weight, in general a very different relative occupation from that outside the crossing region, and this consequently modifies significantly the weights of the network of states with which they interact, directly and indirectly, as described by the rate equations. This is exhibited explicitly for the simple example worked out in Sec. IIIB, where the occupations of all states between those labeled n_1 and n_2 are strongly modified in this regime. On the other hand, if the coupling to the bath is relatively large, $\Gamma \gg \Delta$, then physical intuition suggests that the interaction with the bath effectively broadens the quasienergy dispersion curves by more than the gap itself, such that the avoided crossing should have no consequences on the statistical properties of the system. We have shown that this is indeed the case, namely that the density matrix (in a λ -independent basis) remains virtually unaffected.

Having studied in detail Floquet systems spanned by a finite basis, we have looked at the consequences when the size of the basis set increases without limit, as for a particle confined by an unbounded potential and driven periodically. We have concluded that, in many cases, the important anomalies of such systems, including the appearance of an infinite number of weakly avoided crossings and the non-convergence of the Floquet states with increasing basis size, play no important role for the physical properties of the system in contact with a heat bath, confirming our physical intuition. One can safely employ, in numerical calculations, a finite suitably truncated basis set for such a system. However, a special feature of such a Floquet system, which has a dense quasienergy spectrum, is that its statistical properties depend in general on both the magnitude and the overall strength of the coupling between

the system and the bath.

These calculations have been based on a perfectly periodic driving field. We recognize that effects that we have neglected, notably fluctuations in amplitude and frequency of the driving field, are similarly expected to wash out the effects of sufficiently weakly avoided crossings. Those effects will be examined in a future publication.

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APPENDIX A: VALIDITY OF LOWEST ORDER BORN APPROXIMATION

In the still exact Eq. (2.4) we have made the lowest order Born approximation by using the factorization Eq. (2.5) of the total density matrix directly on the right hand side of that equation. In order to check the validity of this approximation, we analyze in this appendix the fourth order correction arising from the factorization of the total density matrix only in the first iterate of Eq. (2.4), namely

$$\begin{aligned}
& + \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt''' \text{Tr}_b \left[\tilde{H}_{sb}(t), [\tilde{H}_{sb}(t'), [\tilde{H}_{sb}(t''), [\tilde{H}_{sb}(t'''), \tilde{\rho}(t''') \otimes \rho_b]]]] \right] \\
& - \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt''' \text{Tr}_b \left[\tilde{H}_{sb}(t), [\tilde{H}_{sb}(t'), \text{Tr}_b \left([\tilde{H}_{sb}(t''), [\tilde{H}_{sb}(t'''), \tilde{\rho}(t''') \otimes \rho_b]] \right) \otimes \rho_b] \right]
\end{aligned} \tag{A1}$$

Each of these two terms has the structure of three nested time integrals, with a sum of integrands from the nested commutators (the terms differing only by the order of the factors), each of which is a product of 4 system operators and a fourth order correlation function of the bath. A representative such bath correlation function is

$$\langle \tilde{B}(t) \tilde{B}(t') \tilde{B}(t'') \tilde{B}(t''') \rangle - \langle \tilde{B}(t) \tilde{B}(t') \rangle \langle \tilde{B}(t'') \tilde{B}(t''') \rangle \tag{A2}$$

$$= \langle \tilde{B}(t) \tilde{B}(t'') \rangle \langle \tilde{B}(t') \tilde{B}(t''') \rangle + \langle \tilde{B}(t) \tilde{B}(t''') \rangle \langle \tilde{B}(t') \tilde{B}(t'') \rangle \tag{A3}$$

$$= G(t - t'') G(t' - t''') + G(t - t''') G(t' - t''), \tag{A4}$$

where the equality of (A3) to (A2) makes use of the generalized Wick theorem [18]. The final pair of correlation function products (A4) has the important property that in each of

the two terms the time intervals overlap. (The only combination where this is not the case is explicitly subtracted in (A2).) Thus all three times appearing in the integrals, t' , t'' , and t''' , must be close to t in order to give an important contribution, since $G(\tau)$ is significant only for $\tau < \tau_c$. This same condition holds for *all* contributions to the fourth order term. Each such term has an upper limit given by

$$\Gamma_4 \equiv \gamma^4 \tau_c^3 \langle A^4 \rangle \langle B^2 \rangle^2, \quad (\text{A5})$$

which compared to the magnitude of the second order rate Γ_2 is small if the weak coupling condition, $\tau_c \Gamma_2 \ll 1$, Eq. (2.10), is fulfilled, namely that during the bath correlation time τ_c the impact of the second order terms is small; Eq. (2.10) is itself sufficient to ensure the smallness of higher order corrections.

A more restrictive condition, $\Gamma_2 < \varepsilon_{ij}$ for all i, j , was imposed in Ref. [7], namely that the system-bath coupling must be smaller than the smallest quasienergy splitting. In contrast, the above estimate of the fourth order term shows that larger system-bath couplings with $\Gamma_2 > \varepsilon_{ij}$ can equally well be treated with the present approach, as long as the weak coupling condition, Eq. (2.10), is fulfilled.

APPENDIX B: VALIDITY OF TIME AVERAGING OF THE RATES

The replacement of rates by their averages over a driving period (see Eq. (2.19) and the sentence preceding it) relies on the assumption that the density matrix elements do not vary substantially over that period, $T = 2\pi/\omega$. At first glance this seems to be wrong as, e.g., an off-diagonal density matrix $\rho_{ij}(t)$ with an associated quasienergy difference ε_{ij} , that can be as large as ω , would have in the absence of the coupling to the heat bath a substantial variation over the time T . But it will turn out that such off-diagonal matrix elements (corresponding to large quasienergy differences) are in fact small at large times.

We use the observation that the set of differential equations (2.18) is linearly coupled and has time-periodic coefficients $R_{ij;ki}(t)$ with period T . Thus one can apply the Floquet theorem, which requires the general solution to be of the form

$$\rho_{ij}(t) = \sum_n a_n e^{\lambda_n t} r_{ij;n}(t), \quad (\text{B1})$$

with time-periodic functions $r_{ij;n}(t + T) = r_{ij;n}(t)$, and $e^{\lambda_n T}$ an eigenvalue of the time-evolution operator for all density matrix elements over one period. (The λ_n are assumed

here to be non-degenerate.) This solution is in general not periodic in time.

In the following we are interested in the solution for large times. All solutions with $Re(\lambda_n) < 0$ decay to zero in this limit and are of no further interest. From the general properties of a density matrix, $0 \leq \rho_{ii} \leq 1$ and $|\rho_{ij}|^2 \leq \rho_{ii}\rho_{jj} \leq 1$, it follows that solutions with $Re(\lambda_n) > 0$ are impossible. So we are left with discussing solutions with $Re(\lambda_n) = 0$. For the trace of the density matrix to be 1, i.e. time-independent and nonzero, there must be a solution $\lambda_0 = 0$. Thus we have a stationary solution for the density matrix elements

$$\rho_{ij}(t) = r_{ij;0}(t), \quad (\text{B2})$$

which is time-periodic, with at least some diagonal components which have non-zero time averages. (We cannot exclude that in addition to this, there is a traceless quasiperiodic solution from pairs of purely imaginary λ_n . As its time average would be zero, we ignore it in the following.)

We now expand this stationary time-periodic solution in Fourier coefficients

$$\rho_{ij}(t) = \sum_K \rho_{ij}(K) e^{iK\omega t}, \quad (\text{B3})$$

so that the above differential equations (2.18) reduce, for each of the Fourier components, to

$$\begin{aligned} -i(\varepsilon_{ij} + K\omega)\rho_{ij}(K) &= \frac{1}{2} \sum_{k,l,M} \left[\rho_{lj}(K-M) R_{ik;lk}(M) + \rho_{il}(K-M) R_{jk;lk}^*(-M) \right. \\ &\quad \left. - \rho_{kl}(K-M) (R_{lj;ki}(M) + R_{ki;lj}^*(-M)) \right] \end{aligned} \quad (\text{B4})$$

The right hand side is second order in the small system-bath coupling constant γ , because each rate factor (2.15) is. Although we are explicitly concerned with couplings which are not arbitrarily weak with respect to the smallest quasienergy differences in the problem, we do want to consider, as in the conventional static case, coupling which is weak on higher energy scales, such as the driving frequency ω . Then for non-zero values of K the necessary smallness of the left hand side of (B4) must be achieved by the smallness of $\rho_{ij}(K)$, and it is consistent to neglect all such components of the density matrix in the solution; we will set

$$\rho_{ij}(K \neq 0) = 0, \quad (\text{B5})$$

and we will simply write ρ_{ij} for $\rho_{ij}(K = 0)$, which is by definition time independent. (We point out that this implies a time-periodic density *operator* for large times, as one would

expect physically.) This time independence of ρ_{ij} at large times then implies in (B4) that only $M = 0$ terms appear — i.e., that the complex rates R can be replaced by their time averages $R(M = 0)$. (Note that for a quasienergy difference $\varepsilon_{ij} \approx \pm\omega$ between a Floquet state close to the lower boundary and a state close to the upper boundary of the chosen strip $0 \leq \varepsilon < \omega$, one would need to choose $K = \mp 1$ in Eq. (B4) or choose a conveniently displaced strip.)

APPENDIX C: DERIVATION OF THE EFFECTIVE RATE R^{ac}

In this appendix we derive the rate equations for the reduced density matrix elements in the diabatic basis of an isolated weakly avoided crossing in the stationary limit. This leads to an effective rate R^{ac} , which expresses the entire λ -dependence related to an avoided crossing.

Starting from the operator equation (2.8) one follows the steps leading to Eq. (2.20), but now in the diabatic basis of the avoided crossing, which is related to the adiabatic basis by

$$|u_a(t)\rangle = \alpha|u_1(t)\rangle + \beta|u_2(t)\rangle \quad (\text{C1})$$

$$|u_b(t)\rangle = \beta|u_1(t)\rangle - \alpha|u_2(t)\rangle \quad (\text{C2})$$

with $\alpha = \sqrt{(1 + d/\sqrt{1 + d^2})/2}$, $\beta = \sqrt{(1 - d/\sqrt{1 + d^2})/2}$ and d , the dimensionless distance from the center of the avoided crossing, is defined in Eq. (3.2). This basis change alters the left hand side of Eq. (2.20), as the coherent dynamics is less conveniently described in the diabatic basis. The right hand side is left invariant, but now with rates in the diabatic basis, and can be simplified with the following assumptions:

1. We assume that the avoided crossing is sufficiently weak and isolated within the quasienergy spectrum, such that we can neglect any dependence of diabatic states 1 and 2 on the driving amplitude λ , valid over a sufficiently limited range of λ near λ_0 . We assume the same for all other Floquet states, such that the diabatic basis is independent of λ within that range.
2. All other quasienergy separations near $\lambda = \lambda_0$ are assumed to be much larger than the splitting Δ as well as larger than the effective coupling to the bath, such that all off-diagonal density matrix elements can be neglected, except for ρ_{12} and ρ_{21} .

3. We define rates $R_{ij;kl}$ in the diabatic basis in analogy to Eqs. (2.19) and (2.15), where the λ dependence appearing in the quasienergies ε_1 and ε_2 is neglected in the neighborhood of the avoided crossing. That is, the Fourier transformed bath correlation function $g(\varepsilon - m\omega)$ is taken to be constant over this very small range of ε . This is a good approximation as long as $\Delta\tau_c \ll 1$ holds, which is the case for the weakly avoided crossings we are interested in. Together with assumption 1 this makes the rates $R_{ij;kl}$ independent of λ .
4. We set $A_{12}(m) = A_{21}(m) = 0$ (see Eq. (2.13)), as we assume that the spatial structures of states 1 and 2 are very different. We are explicitly looking at a very weakly avoided crossing, and this typically arises from such dissimilar pairs of states, such as those arising from undriven states one of which is of low and the other of very high energy. Similarly, any other state j cannot be spatially similar to 1 and 2 simultaneously, leading to $A_{1j}(m)A_{2j}(m) = 0$.
5. For simplicity we restrict the discussion to periodic drivings that are symmetric, i.e. $H_s(t) = H_s(-t)$. This makes the rate $R_{22;11}$, which appears in the definition of Γ in Eq. (3.5), real.

For the density matrix in the subspace of states 1 and 2 we find

$$\Delta \operatorname{Im}(\rho_{12}) = -\rho_{11} \sum_k R_{1k} + \sum_k \rho_{kk} R_{k1} \quad (\text{C3})$$

$$-\Delta \operatorname{Im}(\rho_{12}) = -\rho_{22} \sum_k R_{2k} + \sum_k \rho_{kk} R_{k2} \quad (\text{C4})$$

$$i\Delta[(\rho_{11} - \rho_{22}) - 2d\rho_{12}] = \rho_{12}\Gamma \quad (\text{C5})$$

with the symbol Γ introduced in Eq. (3.5).

The third equation expresses the off-diagonal element ρ_{12} in terms of the diagonal elements, as explicitly written in Eq. (3.6). Consequently, the left hand side of the first two equations can be written in terms of the diagonal elements,

$$\Delta \operatorname{Im}(\rho_{12}) = R^{\text{ac}}(\rho_{11} - \rho_{22}) \quad (\text{C6})$$

where we use the notation R^{ac} introduced in Eq. (3.4). We now redefine the effective rates \bar{R}_{ij} in the diabatic basis, equal to the original rates R_{ij} except for $\bar{R}_{12} = R^{\text{ac}}$ and similarly $\bar{R}_{21} = R^{\text{ac}}$ (note $R_{12} = 0$ by assumption 4 above). This allows us to rewrite the first two

equations in the standard rate equation form, as if there were just diagonal density matrix elements: Eq. (3.3).

APPENDIX D: DIAGONALIZING THE DENSITY MATRIX

It is instructive to ask in which basis the reduced density matrix is diagonal near a weakly avoided crossing, and whether this is close to the diabatic 1,2 basis above, or close to the a, b basis of the Floquet states. The relevant sector of the density matrix in the 1,2 basis is given by

$$\rho = \begin{pmatrix} \rho_{11} & A(\rho_{11} - \rho_{22}) \\ A^*(\rho_{11} - \rho_{22}) & \rho_{22} \end{pmatrix}, \quad (\text{D1})$$

with

$$A = \frac{1}{2d - i\Gamma/\Delta}. \quad (\text{D2})$$

It is diagonal in the basis of the two states

$$\begin{aligned} |u_{d1}\rangle &= \left[\frac{A}{|A|} \sqrt{1 + \frac{1}{z}} |u_1\rangle + \sqrt{1 - \frac{1}{z}} |u_2\rangle \right] / \sqrt{2} \\ |u_{d2}\rangle &= \left[-\frac{A}{|A|} \sqrt{1 - \frac{1}{z}} |u_1\rangle + \sqrt{1 + \frac{1}{z}} |u_2\rangle \right] / \sqrt{2}, \end{aligned} \quad (\text{D3})$$

with

$$z = \sqrt{1 + 4|A|^2}, \quad (\text{D4})$$

leading to diagonal elements

$$\rho_{d1,d1} = [\rho_{11}(1 + z) + \rho_{22}(1 - z)]/2 \quad (\text{D5})$$

$$\rho_{d2,d2} = [\rho_{11}(1 - z) + \rho_{22}(1 + z)]/2. \quad (\text{D6})$$

One can check that this gives the expected result in two limiting situations:

- Far from the avoided crossing, $d \gg 1$, or for $\Gamma \gg \Delta$, the effective rate R^{ac} between states 1 and 2 becomes small, we have $A \rightarrow 0$ and $z \rightarrow 1$, and we regain the 1,2 basis as that in which the reduced density matrix is diagonal.
- For weak coupling, $\Gamma \ll \Delta$ and near the avoided crossing, $d \ll 1$, we have $z \gg 1$. It is immediately clear that at the center, where $d \rightarrow 0$, the appropriate basis states for a

diagonal density matrix become nearly the equal admixtures of states 1 and 2, which are the Floquet states a and b . In fact, this is the case for all d in this limit of weak coupling to the bath; for infinitesimal Γ these states which diagonalize ρ are *exactly* the Floquet states.

We can readily understand these two limits in terms of the balance between energy pumping of the system by the periodic field and relaxation by the bath. For $\Gamma \ll \Delta$ the near resonant periodic driving dominates, and the Floquet states in the absence of the bath are approximate solutions, only weakly relaxed. In the opposite limit, $\Gamma \gg \Delta$, those Floquet levels are effectively broadened more by the bath interaction than their separation, and the mixing of the diabatic states is a perturbation on their relaxation by the bath.

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